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INVESTIGATION OF THE MECHANISMS ASSOCIATED WITH GAS BREAKDOWN UNDER INTENSE OPTICAL ILLUMINATION

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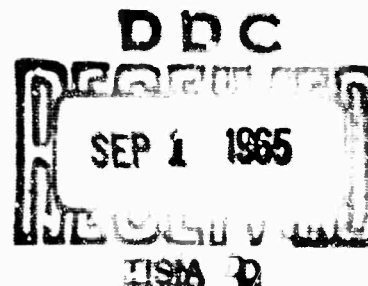
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SUMMARY

The Research Laboratories of the United Aircraft Corporation are conducting under the subject contract an experimental and theoretical investigation of the physical mechanisms involved in the electrical breakdown of gases by intense optical radiation. The focused high-intensity optical frequency beam from a Q-spoiled ruby or neodymium laser is used to cause electrical breakdown in a test gas, and the ionization produced is examined as a function of the gas, pressure, volume within which the breakdown occurs, and frequency of radiation. With both ruby and neodymium radiation, for the gases studied, breakdown in air was observed to require the highest field strength with successively lower field strengths required for breakdown in neon, helium, and argon.

Studies have been carried out to examine the attenuation of the laser beam by the breakdown plasma. For beam intensities slightly above the breakdown threshold, it is observed with both ruby and neodymium incident radiation that more than half of the laser beam energy can be absorbed in the plasma produced by the breakdown and that over 90% attenuation of the laser beam can occur at later times in the optical pulse.

Measurements of the attenuation of an optical beam by the breakdown plasma at times subsequent to the incident giant pulse have been made using the cw beam from a helium-neon laser. These measurements show that the same 90% absorption observed with the incident pulse is obtained with a cw beam for times of the order of milliseconds after the formation of the plasma. This indicates that the breakdown plasmas produced have an extremely long lifetime and that losses are small in the afterglow plasma.

Measurements have been made to examine the effects of diffusion-like losses on the breakdown threshold by varying the focal volume within which the breakdown is formed. For all of the gases studied it was observed that the breakdown threshold

electric field strength with both ruby and neodymium radiation is inversely related to the dimensions of the breakdown region; i.e., breakdown within small focal volumes requires a larger optical frequency electric field than is necessary for larger volumes. These measurements have been made for gas pressures varying from atmospheric to 2000 psi, and at all pressures the same effect is noted. The observed volume dependent breakdown threshold implies that even at pressures as high as 2000 psi diffusion-like losses are a dominant mechanism in the development of optical frequency breakdown, a result not considered in existing theories of the optical breakdown process.

In the experiments with neodymium irradiation, for the larger focal volumes with both helium and argon a pronounced minimum is present in the breakdown electric field vs. pressure curves. With ruby irradiation no minimum is observed for helium and that for argon is less distinct and shifted to higher pressures. Using the breakdown data obtained with ruby and neodymium laser irradiation, the frequency dependence of the breakdown threshold was evaluated. For either argon, helium, or air the lower frequency neodymium radiation gives a lower breakdown threshold than with ruby at low pressures. At high pressures the neodymium data approaches (helium) or crosses (argon) that for ruby as a result of the minimum observed with neodymium; for air the ratio of the neodymium to ruby breakdown threshold remains approximately constant over the pressure range studied.

Theoretical studies have been carried out which show that existing classical models of the breakdown process are not adequate to explain the phenomena observed at optical frequencies. Multiple photon theories recently proposed are unable to predict the magnitude of the E field required for breakdown or the pressure and volume dependence obtained experimentally. Calculations of the inverse bremsstrahlung cascade theory of the breakdown process have been carried out for optical frequencies where the photon energy is greater than the classically calculated electron oscillation energy and show that an electron exchanges energy with the applied electromagnetic field in increments of the photon energy. This result differs in kind from that obtained using the classical microwave theory and offers an experimental test to examine the validity of the inverse bremsstrahlung model.

INTRODUCTION

The interaction of extremely high-intensity optical frequency electromagnetic radiation with gas atoms has been accessible for experimental study only with recent development of high-powered lasers. Studies of gas breakdown by optical frequency radiation were initiated at the United Aircraft Research Laboratories early in 1962. These Corporate-sponsored investigations demonstrated that breakdown at optical frequencies occurs in many gases at field strengths which can be easily achieved by optical lasers. A pulsed ruby laser was used for the initial studies, and the optical field strength required for breakdown was investigated as a function of gas pressure in argon and helium.

It was determined that field strengths of the order of 10^7 volts per centimeter were required for breakdown and that electron densities greater than 10^{17} electrons per cm^3 were produced in the resulting breakdown plasma. Under the joint sponsorship of the Advanced Research Projects Agency, the Office of Naval Research, and the Department of Defense [Contract Nonr-4299(00)], gas breakdown studies in air were undertaken. The field strengths required for breakdown in air were larger even than those required for helium or argon, indicating that gases of a molecular nature possess energy loss mechanisms in addition to those characteristic of monatomic gases. It was also observed that a significant attenuation of the optical beam, in many cases exceeding 50%, may be produced by the breakdown plasma formed. Direct electron stripping, single or multiple photon absorption, Compton collisions, or conventional microwave breakdown theory do not adequately account for the breakdown phenomena observed, and it was proposed that the quantum mechanical process of inverse bremsstrahlung is the mechanism responsible for breakdown at optical frequencies. These results have been reported in the Proceedings of the Sixth International Conference on Ionization Phenomena in Gases, Paris, France, July 8-13, 1963; Physical Review Letters, Vol. 11, No. 9, November 1963; Physical Review Letters, Vol. 13, No. 1, July 1964; and the Final Report C920088-2, under Office of Naval Research Contract Nonr-4299(00).

Under the present subject contract, further studies of the threshold electric field strength required for breakdown as a function of pressure in different gases, the effect of focal spot size on threshold field strength, the frequency dependence of the breakdown threshold, and the attenuation of the laser radiation by the breakdown plasma are being conducted to more fully define the nature of the breakdown mechanisms at optical frequencies.

RESEARCH PROGRAM

Apparatus

The high-intensity light source used in these studies is a Q-spoiled laser beam produced by the laser system shown schematically in Fig. 1. Both ruby and neodymium lasers have been employed in order to examine the frequency dependence of the breakdown. The Brewster-angled laser rod, either ruby or neodymium, is positioned in an optical cavity formed by the two dielectric mirrors, and a Kerr cell-polarizer shutter is used to alter the cavity Q to produce a single giant pulse of approximately 20 Mwatts peak power from the laser system. The output laser beam is focused by a lens which forms one window of a cell containing the test gas. The test cell, constructed of stainless steel with Teflon gaskets, is designed for both vacuum and high-pressure use and permits the study of a variety of gases under high-purity conditions at pressures up to 2000 psi. A set of charge collection electrodes is located within the test cell to measure the ionization produced in the breakdown, and

aperture windows are provided in the cell to permit both visual observation of the breakdown and photomultiplier measurements of the breakdown luminosity time history. An extension port was incorporated into the high-pressure cell to accommodate different focal length lenses for the studies of the focal volume dependence of the breakdown.

In order to interpret the results obtained, measurements have been made of the output giant pulse shape, energy, and beam divergence. An ITT photodiode with a 0.35 nsec rise time was used to observe the time history of the laser pulse. The output from the diode was fed directly into a Tektronix 519 oscilloscope, giving an over-all system response time of 0.45 nsec. A photograph of a typical output pulse from the ruby and neodymium laser systems is shown in Fig. 2. Measurements with a high-sensitivity photomultiplier system having a response time of less than 2 nsec were carried out using a much slower oscilloscope sweep speed, and adjustments made in the operating conditions of the laser such that any output other than the single giant pulse was at a power level less than 1% of the giant pulse. This was done to insure that the breakdown under study was the result of the single giant pulse and not the accumulated effect of a sequence of pulses. As seen in Fig. 2, both the ruby and the neodymium giant pulses have a roughly triangular shape with half-widths of 20 and 50 nsec, respectively.

The calibration system shown in Fig. 1 was used to measure the laser giant pulse energy and wave shape for every shot. Partially reflecting mirrors reflect a portion of the output beam onto the photodiode and a calorimeter to determine the pulse time history and beam energy. The calorimeter reading was calibrated in terms of the transmitted beam energy by the use of a second reference calorimeter located directly in the transmitted output beam. The laser beam power was then calculated for each shot from these measurements of laser time history and total energy.

The laser beam output in the giant pulse mode is not fully collimated but, in fact, has a small divergence. For the laser beam diameters used in these experiments, the focusing is not diffraction limited, and from geometric optics, a lens will focus this slightly divergent beam to a focal diameter given by the product of the lens focal length and angle of divergence, $d = f\theta$. The divergence is first determined from the diameter of the focal spot produced with an extremely long focal length lens. For these measurements, the laser beam is attenuated to the point that the focused beam intensity is just below that necessary to record on a piece of exposed polaroid film. By then removing an attenuation of 50% from the beam, the signal at every point is doubled and the half-power point of the incident focused beam is at the recording threshold of the film. The diameter of the film image is then the diameter at half power of the focused laser beam. From the half-power diameter and the lens focal length, the laser beam divergence can then be calculated. Using this procedure, beam divergences of 4.5 milliradians and 3.0 milliradians were obtained for the ruby and neodymium lasers, respectively. From the laser pulse power and divergence and the focal length of the lenses used in the breakdown experiments, the beam power density of the radiation was determined, and using Poynting's theorem, the optical frequency electric field at the lens focus was calculated.

Charge Collection Studies

A pair of charge collection electrodes, shown in Fig. 1, were located within the high-pressure cell on either side of the breakdown region and were used to measure the ionization produced in the breakdown. The electrical circuit used for these measurements is shown in Fig. 3. A 90-volt potential source, shunted by a series of capacitors for high-current response, was connected to each of the electrodes, one positive and the other negative. Using a type Z differential amplifier with a Tektronix 551 dual beam oscilloscope, the number of electrons and ions collected, respectively, at the positive and negative electrodes were observed across the 1 microfarad capacitors shown in the circuit. As shown in Fig. 3, electrons and ions of the order of 10^{13} ion pairs were collected from the breakdown plasma at atmospheric pressure. The charge collected was observed to decrease with increasing pressure, most likely due to increased recombination at the higher pressure. From Fig. 3, electron and ion collection occur simultaneously, indicating that free diffusion is not important in the charge separation process.

Absorption by a Breakdown Plasma

In the course of the gas breakdown studies carried out under Contract Nonr-4299(00), it was observed that when breakdown occurs the transmitted laser radiation is severely attenuated during the later portions of the laser optical pulse. Studies of this attenuation were carried out, and it was established that the energy removed from the optical beam was not scattered at the laser frequency or reradiated by excited atoms but instead was truly absorbed by the breakdown plasma.

Under the present contract, investigations have been made of the attenuation of 1.06μ neodymium laser radiation by the breakdown plasma using the apparatus shown in Fig. 4. Photomultiplier A, shown in the figure, was used to observe the transmitted radiation. The monitor photomultiplier (B) was used to determine that the incident laser pulse remained constant for each firing and was not affected by the occurrence of breakdown. When no breakdown occurred, the transmitted beam had the time history of the upper trace in Fig. 4, a wave shape identical to that observed by photomultiplier (B). When breakdown did occur, while the wave trace of the monitor photomultiplier remained unchanged, the transmitted radiation observed by photomultiplier A was severely attenuated after the first 6 to 10 nsec, as shown by the lower trace in Fig. 4. For these experiments the beam power is slightly above the breakdown threshold for the argon test gas, and over one half of the one-joule incident optical energy is removed from the transmitted beam. At times shortly after the initiation of the breakdown, approximately 90% of the incident radiation is being withdrawn from the transmitted beam by the plasma, a result which parallels that observed previously with ruby radiation.

As shown in Fig. 4, during the later portions of the incident giant optical pulse, less than 10% of the incident radiation is transmitted through the breakdown plasma. Measurements have been made of the attenuation of $.6328 \mu$ helium-neon laser

radiation by the breakdown plasma to examine this effect at times subsequent to the incident giant laser pulse. Using the optical configuration shown in Fig. 5, the helium-neon laser beam was directed through the breakdown plasma at right angles to the incident giant pulse beam and the transmitted fraction of the helium-neon beam detected by a photomultiplier to record only $.6328 \mu$ radiation. A series of measurements were carried out with the breakdown plasma produced by the neodymium laser pulse in argon at a series of pressures from atmospheric pressure to 500 psi. The results obtained at 480 psi are as shown in Fig. 9. When breakdown occurs the helium-neon beam is attenuated by the same 90% as the incident giant pulse, and the attenuation was observed to persist for a time of several milliseconds compared with the 25 nsec duration of the initiating giant laser pulse. This result, coupled with framing camera photographs of the breakdown luminosity, shows that the breakdown plasma has an extremely long lifetime and indicates a low rate of energy loss from the afterglow plasma. In many cases severe fluctuations in the attenuation were observed during the absorption of the helium-neon beam by the afterglow plasma. In some instances this absorption may decrease to nearly zero for a tenth of a microsecond and then return to the earlier value of 80 to 90%. The cause of these fluctuations is not fully understood but may be related to either reflection of the helium-neon beam by the plasma region or changes in the shape of the afterglow plasma as a function of time. Further studies of these effects will be carried out coupling the helium-neon beam attenuation measurements with framing camera measurements of the growth and configuration of the initial breakdown and of the afterglow plasma.

Pressure Dependence of Breakdown Threshold

In the course of the charge collection experiments, it was observed that below a certain threshold laser power neither charge production nor the visible discharge at the lens focus was observed and that this breakdown threshold changed with pressure. A systematic study of the pressure dependence of the breakdown threshold was carried out in air, argon, helium, and neon over pressures ranging from atmospheric pressure to 2000 psi. The results of these studies with both ruby and neodymium laser irradiation are shown in Fig. 6. In the figure, the strength of the optical frequency electric field of the focused laser beams at threshold is plotted as a function of pressure for the several gases. The rms electric field was obtained by equating the Poynting theorem expression for electromagnetic energy flow to the experimentally determined laser power flux in the focused beam; i.e., $\epsilon E_{rms}^2 c \pi f^2 (\frac{\theta}{2})^2 = \frac{E}{\tau}$, where E is the energy and τ the half-width of the laser pulse. As observed in Fig. 2, the giant pulse for both the ruby and neodymium lasers are roughly triangular in shape, and the rms E field thus determined is that associated with peak laser beam intensity.

The data shown in Fig. 6 was obtained using a 3 cm focal length lens in the high-pressure cell (see Fig. 1), and for each of the gases studied, the breakdown threshold is observed to decrease with pressure leveling off at the higher pressures. Argon has the lowest ionization potential (15.7 eV) of the gases studied and, with both ruby and neodymium irradiation, required the least electric field for breakdown. Neon and helium with ionization potentials of 22 and 24 eV, respectively, have substantially higher breakdown thresholds, with that for neon lying slightly above the threshold for

helium. This result differs from that obtained previously by Tomlinson;¹ however, his recently reported results² show the higher breakdown threshold for neon as obtained here.

Focal Volume Dependence of Breakdown

To determine whether the development of breakdown is controlled by the balance between losses associated with the small volume of the breakdown region and the rate of energy addition or limited only by the rate of addition of energy by the focused beam, experiments were carried out to examine the focal volume dependence of the breakdown threshold. A breakdown threshold independent of volume would indicate that only the radiation interaction is important, while a volume dependence would show the presence of diffusion-like losses which must be taken into account in the theoretical interpretation of the results obtained.

Experimentally, the plasma is initially formed within the region of the intense focused beam, a cylindrical volume of diameter, $d = f\theta$, and length, $l = (\sqrt{2}-1)\frac{f^2\theta}{D}$, where f is the lens focal length, θ the laser beam divergence, and D the laser beam diameter. The volume of the focal region is then given by $V = \frac{\pi}{4} \frac{f^4\theta^3}{D} (\sqrt{2}-1)$, the boundaries of this volume corresponding to the half-power points of the focused radiation. To test the effects of diffusion-like losses on the development of breakdown, measurements were made of the breakdown threshold as a function of the initial plasma formation volume. Since the laser beam divergence and diameter remained constant from pulse to pulse, the focal volume was conveniently changed by using different focal length lenses to focus the laser radiation. The experiments to examine the focal volume dependence of the threshold were carried out using the neodymium and ruby laser pulses to produce breakdown in argon, helium, air, and neon at a series of pressures from atmospheric pressure to 2000 psi using lenses with focal lengths from 3 cm to 15 cm. The results obtained are shown in Figs. 7, 8, and 9. A characteristic length associated with diffusion-like losses is the diffusion length, Λ , which for a cylindrical volume is given by $\frac{1}{\Lambda^2} = \left(\frac{\pi}{l}\right)^2 + \left(\frac{2.4}{r}\right)^2$, and the breakdown thresholds for argon, helium, air, and neon are shown in Fig. 7 as a function of this parameter. As indicated above, if diffusion-like losses are not important in the development of the breakdown, the threshold should be independent of Λ . It is apparent from the figure, however, that this is not the case; breakdown within small focal volumes (small Λ) requires several times the threshold field strength as for the larger volumes studied. From Fig. 7, the breakdown varies with the diffusion length as $E \propto \Lambda^{-\frac{3}{4}}$ over the range of focal lengths used, and experimentally the slope is the same for all of the gases studied. Some leveling off of this slope is observed for the largest focal volumes, and further studies are planned to extend the measurements to still larger diffusion lengths. The results obtained indicate that diffusion-like losses are definitely involved in the development of breakdown, and from the finite slope of the curves, the loss-free ($\Lambda \rightarrow \infty$) breakdown threshold for the gases studied lies at still lower electric field strengths. As a result, the magnitude and nature of the diffusion-like losses must be known in order to develop an adequate theoretical prediction of the experimental breakdown threshold.

The breakdown threshold due to neodymium and ruby laser irradiation as a function of pressure for different focal length lenses are shown in Figs. 8 and 9 for helium, argon, and air, the curves labeled by the appropriate diffusion length. It is observed experimentally that with neodymium irradiation for the larger focal volumes with both helium and argon a pronounced minimum is present in the breakdown electric field vs. pressure. With $\Lambda = 9.2 \times 10^{-3}$ cm, the threshold minimum occurs at approximately 5×10^4 mm Hg for helium and for argon at 2.5×10^4 mm Hg. No minimum is apparent in the breakdown data for the shorter focal length lenses. For air the minimum is not so distinct and, if present, lies at still higher pressures. With ruby irradiation, no minimum is observed in either the air or helium breakdown threshold curves, and that for argon is less distinct and is shifted to higher pressures ($\sim 7 \times 10^4$ mm Hg).

In the course of these experiments it was observed that the breakdown location for the larger focal length lenses occurred at a distance downstream from the focus, particularly at the highest pressures. For example, with the 15 cm focal length lens at a gas pressure of 2000 psi, the breakdown occurred at approximately 17 cm from the lens. It was also observed that with the long focal length lenses frequently more than one breakdown developed along the beam axis. Further studies of these phenomena are being undertaken to determine their causes and their effect on the development of breakdown.

Frequency Dependence of Breakdown

From the breakdown data obtained with the ruby and neodymium laser irradiation, the frequency dependence of the breakdown threshold can be obtained. The ruby and neodymium laser beams have different divergences, and in determining the frequency dependence, the breakdown threshold must be compared for equal values of the diffusion length parameter, Λ . Shown in Fig. 10 are the breakdown thresholds for argon, helium, and air with ruby and neodymium irradiation for a Λ of 2.4×10^{-3} cm. The lower frequency neodymium pulse gives a lower breakdown threshold than for ruby at low pressures, the ratio between the thresholds being approximately 1.8 at atmospheric pressure. At high pressures the neodymium data approaches (helium) or crosses (argon) that for ruby as a result of the minimum observed with neodymium. For the breakdown of air, as shown in Fig. 10, the ratio of the neodymium to ruby breakdown threshold remains approximately constant over the pressure range studied.

THEORIES OF OPTICAL FREQUENCY BREAKDOWN

A number of papers have recently been published proposing theories to explain the electrical breakdown of gases by optical frequency radiation. For completeness, both these theories and those proposed earlier^{3,4} will be discussed in order to evaluate the present status of the theoretical understanding of optical frequency breakdown.

Direct Electron Stripping

From Figs. 6 to 10, the breakdown threshold optical frequency electric fields range from 10^6 to 10^7 volts per cm depending on gas and pressure, and such high fields might distort the coulomb atomic field sufficiently that the electron is no longer bound. For hydrogen the binding field experienced by the ground state electron is $E = \frac{e}{a_0^2}$, where a_0 is the radius of the first Bohr orbit. This field is 5.8×10^9 volts per cm, and it is evident that the breakdown field strength of even 10^7 volts per cm are not large enough to result in direct electron stripping.

At some atomic radius, however, the applied field is greater than the coulomb binding field, resulting in a potential maximum with a decrease in potential at still greater radii. At only 10^{-6} cm from the nucleus, the potential is such that the ground state electron may tunnel through this potential maximum. (10^{-6} cm is considerably less than the wavelength of the radiation used, so that the uniform field approximation is a valid one.) Estimates of the rate of ionization by this process have been made by Bunkin and Prokefov,⁵ and their calculations show that field strengths of several times 10^8 volts per cm would be required. These field strengths are one to two orders of magnitude larger than determined experimentally for breakdown, and this, therefore, does not appear to be the rate-limiting process in the breakdown.

Compton Collision Energy Transfer

Electrons in the gas can receive energy from the incident radiation by Compton collisions with photons in the laser beam. Averaging over all angles the differential Compton cross section times the energy transfer per collision for the low-energy photons ($h\nu \ll mc^2$) considered here, the power transferred to an electron by this process is given by

$$P_c = \frac{3}{4} r_0^2 h\nu \frac{h\nu}{mc^2} C \frac{E_{rms}^2}{h\nu}$$

where r_0 is the classical electron radius. For the conditions of breakdown P_c is approximately 2×10^{-1} eV/sec electron. The breakdown occurs in a time of the order of 10^{-8} sec, and an energy transfer of 2×10^{-9} eV per electron is certainly not sufficient to cause the ionization of gases whose ionization potentials range from 13 to 24 eV.

Photoionization and Multiple Photon Absorption

The energy of the ruby and neodymium laser photons is 1.78 eV and 1.17 eV, respectively. The gases studied in the breakdown experiments require from 13 to 24 eV for ionization and from 10 to 20 eV even for excitation. As a result, it is not possible for direct photoionization to cause the ionization observed.

It is possible that an atom could absorb either sequentially or simultaneously a number of photons and become ionized by some form of multiphoton process. In

sequential multiphoton absorption, an electron initially in the ground state is raised to a virtual state which persists for a time of the order of 10^{-12} to 10^{-13} seconds. During this time a second photon can be absorbed, raising the atom to a still higher virtual state. By a succession of such absorptions the electron could reach the first excited state from which, by further multiple photon absorptions or photoionization to higher energy levels, the ionized state is eventually reached. However, to reach even the lowest lying levels of argon and helium would require, respectively, 7 and 12 successive photon absorptions. Assuming even a 10% probability for each absorption process, this would imply a 10^5 order difference between the intensities of the laser radiation required for breakdown in argon and helium. The experimentally observed ratio of laser beam intensities is only a factor of three, and on this basis, sequential multiple photon absorption will not account for the development of optical frequency breakdown.

Gold and Bebb⁶ have carried out a semi-classical calculation of multiphoton absorption in the noble gases. They calculate the transition probability for the simultaneous absorption of the number of photons required to ionize the atom and under ruby irradiation for Xe, Kr, Ar, Ne, and He require the absorption of 7, 8, 9, 13, and 14 photons, respectively. The predominant terms in their calculated transition probabilities are the contribution of high-order "near resonant" absorption steps. It is pointed out in the paper that, because of the high-order dependence of the absorption on photon flux for such processes, even gross errors in the matrix elements calculated will not materially affect the predicted breakdown threshold. The calculated breakdown thresholds, however, are three orders of magnitude larger than obtained experimentally. Gold and Bebb argue that photon fluxes several orders of magnitude larger than the average measured value may be present in the beam due to multiple axial mode mixing. Due to the different divergences associated with such axial modes and the aberrations of the simple spherical lenses used, the different axial modes will not be preserved at the lens focus but instead will be smeared out within the focal volume. The smeared-out modes can locally interfere in the focused beam, but such interference is more accurately described as fluctuations in the local photon density. Such fluctuations, for an interference type phenomenon, occur within a volume of the order of the wavelength cubed. Because of the large photon density in the focused beam, in a volume of λ^3 these fluctuations are less than one per cent of the average photon density, and it would appear that the intensity variations of two to three orders of magnitude suggested in the article by Gold and Bebb do not occur. Because of a set of strongly coupled levels in neon, Gold and Bebb calculate that neon should break down more easily than argon even though its ionization potential is considerably greater than that of argon. The ratio they calculate for the threshold electric field required for neon to that of argon is approximately 0.8. As shown in Fig. 6, neon experimentally has a higher breakdown threshold than argon over the entire pressure range studied (of the order of 1.7 times greater), and it does not appear that the multiphoton theory of Gold and Bebb is a valid description of optical frequency breakdown.

Tozer⁷ has also proposed a multiphoton absorption theory in which the statistics of photon density fluctuations in the irradiating beam are taken into account. This

differs from the situation proposed by Gold and Bebb in that the fluctuations considered here are not within a volume of λ^3 resulting from random mode mixing in the beam but instead are the statistical variation in the number of photons within the interaction volume of the atom. The interaction volume is approximated as a cylinder of end area σ , the multiphoton absorption cross section of the atom, and length ct , where $t \sim \frac{h}{h\nu} \sim \frac{1}{\nu}$ the uncertainty time for an interaction involving energy $h\nu$. For the experimental breakdown beam intensity, any reasonable choice of σ gives a number less than one for the number of photons within the interaction volume whose fluctuations therefore can be quite large. Tozer considers the effects of such fluctuations on the simultaneous absorption of sufficient photons to directly ionize an atom and, on the basis of his calculations, predicts both a pressure and a volume dependence for the breakdown. At high pressures Tozer calculates that the breakdown threshold field strength should vary with pressure as $E \propto P^{-1/N_\nu}$, where N_ν is the number of photons which need to be absorbed simultaneously to raise the atom to the first excited state. The predicted threshold monotonically decreases with pressure and offers no explanation for the increasing breakdown threshold observed experimentally (see Fig. 9). Tozer's theory predicts a volume dependence of the breakdown electric field strength of the form $E \propto V^{-1/2N_\nu}$. For helium irradiated with a neodymium laser beam, the predicted volume dependence would then be $E \propto V^{-0.06}$. Experimentally, from the original data used in plotting Figs. 8 and 9, the volume dependence of the breakdown threshold was determined to be $E \propto V^{-0.19}$, a result which differs by a factor of three in the exponent from that predicted. On the basis of discrepancies between the predicted and the experimental pressure and volume dependence of the breakdown threshold, it would not seem that the multiphoton theory offered by Tozer gives an adequate account of the optical frequency gas breakdown.

Microwave Breakdown Theory

Cascade theories have been developed to explain gas breakdown at microwave frequencies⁸ and employed by some to explain optical frequency breakdown.⁹ In the microwave theory, free electrons gain energy from the applied electromagnetic field as a result of elastic collisions between the electrons and the atoms or ions of the breakdown gas. These collisions randomize the ordered oscillatory motion of the electrons in the electromagnetic field, resulting in a net increase in the electron temperature. When they attain sufficient energy, the electrons make inelastic ionizing collisions with the gas atoms, producing a second generation of electrons. The increased electron population again gains energy from the applied field and produces additional generations, the cascade process, for sufficiently high field strengths, ultimately leading to electrical breakdown of the gas.

The rate of gain of energy by an electron in an electric field, E , is $P = eEv$. Solving for the classical motion of an electron in a sinusoidal electric field of amplitude E and frequency ω , the average power absorbed by an electron from the field is

$$\bar{P} = \frac{e^2 E^2}{2m_e m} \left[\frac{\nu m^2}{\nu m^2 + \omega^2} \right]$$

where ν_m is the momentum transfer collision frequency. For a given breakdown volume, at low pressures the dominant loss process is diffusion and at high pressures is elastic energy transfer to the gas atoms. Assuming that only these two loss processes are important, the rate of energy gain by an electron is given by

$$P_{\text{net}} = \frac{e^2 E^2}{2m} \frac{\nu_m}{\nu_m^2 + \omega^2} - \frac{D}{\Lambda^2} \bar{U} - \frac{2m}{M} \bar{U} \nu_m$$

where D is the appropriate diffusion coefficient, M is the mass of a gas atom, and \bar{U} is the average electron energy. For a cw applied field, breakdown will occur for $P_{\text{net}} > 0$, and the breakdown threshold occurs at $P_{\text{net}} = 0$. At low pressures where $\nu_m \ll \omega$, if only diffusion is important the breakdown condition becomes

$$E = \frac{\omega}{\Lambda \nu_m} \left(\frac{2}{3} \bar{U} U_i \right)^{1/2}$$

At higher pressures, the electron energy is dissipated primarily in elastic collisions with the gas atoms, and the cw breakdown threshold occurs for

$$E = \frac{2m}{e} \left(\frac{\bar{U}}{M} \right)^{1/2} (\nu_m^2 + \omega^2)^{1/2}$$

For many gases the momentum transfer collision frequency can be represented as a constant times the gas pressure; i.e., $\nu_m = C_p$. Using this relationship, the cw threshold condition at low and high pressures becomes

$$E = \begin{cases} \frac{1}{\Lambda p} \frac{\omega}{c} \left(\frac{2}{3} \bar{U} U_i \right)^{1/2} & \text{low pressures} \\ (C^2 p^2 + \omega^2)^{1/2} \frac{2m}{e} \left(\frac{\bar{U}}{M} \right)^{1/2} & \text{high pressures} \end{cases}$$

This cw theory gives for the breakdown threshold as a function of pressure the decreasing E field at low pressures and increasing E field at high pressures observed experimentally (the increasing E field definitely observed thus far only with Nd^{+3} irradiation). Similarly, the theory predicts the decreasing breakdown threshold with diffusion length although the dependence observed experimentally is $E \propto \Lambda^{-3/4}$ as compared with the $E \propto \Lambda^{-1}$ predicted.

The irradiating field, however, is not cw but is pulsed with a duration of less than 100 nsec. On this time scale, for the gases and average electron energies expected during the breakdown, electron diffusion, even free diffusion, is not an important process; i.e., few electrons are lost from the breakdown volume during the time of its formation. Thus, even at low pressures, for a pulsed E field the breakdown threshold should be independent of breakdown volume for the conditions of these experiments. From Fig. 7, this is definitely not the case, and it appears that some form of diffusion-like loss other than simple electron diffusion is limiting the breakdown.

If it is assumed that no losses occur and that all of the energy imparted to the electrons from the irradiating field goes into ionization, the energy input rate calculated from the microwave theory could account for the ionization observed and the development of breakdown for the small breakdown volume cases. However, from Fig. 7, it is evident that the diffusion-like losses present are not insignificant and must be considered in evaluating the net energy input to the electrons. Over the range of focal lengths studied, these diffusion-like losses are still important at 240 psi and diffusion lengths of 1.5×10^{-2} cm, and from the decreasing curve of Fig. 7, the infinite volume or diffusion-like loss-free breakdown threshold for helium lies at an E field substantially below the 1.3×10^6 volts per cm measured for these conditions. By comparison, the loss-free microwave calculations give a breakdown threshold of 2.4×10^6 volts per cm and is evidently not a valid model for optical frequency breakdown.

There is, in addition, a serious objection to the application of microwave breakdown theory at optical frequencies. In the microwave theory, the electron between collisions oscillates in response to the applied electromagnetic field. The electron velocity then has the form

$$V(t) = V_0 + \frac{eE}{m\omega} (\cos \delta - \cos \omega t)$$

where δ is the initial phase angle of the field; i.e., the phase angle of the field at the previous collision. The maximum change in velocity between collisions is then

$$V_{\max} = V_0 + 2\left(\frac{eE}{m\omega}\right)$$

giving a maximum energy change of an electron between collisions of

$$\Delta \mathcal{E}_{\max} = 2mV_0 \left(\frac{eE}{m\omega}\right) + \frac{1}{2}m \left(\frac{e^2 E^2}{m^2 \omega^2}\right)$$

For an electron with energy sufficient to cause ionization in argon and an applied field equal to the breakdown threshold, at microwave frequencies $\Delta \mathcal{E} \sim 2 \times 10^{-1}$ eV. Microwave photon energies are of the order of 10^{-5} to 10^{-6} eV, and thus the electron absorbs and emits many microwave quanta per cycle and the electron motion may properly be considered classically. At optical frequencies, for field strengths which result in breakdown, $\Delta \mathcal{E}$ is again $\sim 2 \times 10^{-1}$ eV, but the photon energy in the case of a ruby laser is 1.78 eV. The change in the electron energy between collisions is, in this case, a small fraction of the unit available energy, and it is more reasonable to describe the interaction between the electrons and the optical frequency field by a quantum picture.

Inverse Bremsstrahlung

During an atomic collision an electron may emit a photon by the process of bremsstrahlung. From detailed balance considerations the reverse reaction, inverse

bremsstrahlung, can also occur in which an electron during a collision with an atom or an ion in the gas absorbs a photon of energy from the radiation field. Calculations of this inverse bremsstrahlung process, first proposed to explain optical frequency gas breakdown in Ref. 3, have been carried out as part of a parallel Corporate sponsored program by Professor L. Brown and Dr. P. Mallozzi of Yale University, acting as consultants to the Research Laboratories. These calculations show, under conditions where the photon energy is greater than the classically calculated electron oscillation energy, that during a collision an electron exchanges energy with the applied electromagnetic field in increments of the photon energy. This result differs in kind from that obtained above using the classical microwave theory as applied to optical frequency and is important in developing an understanding of the physics of the optical frequency breakdown process.

The calculations which have been carried out have been made for conditions where the collision frequency of the electrons in the gas is much less than the radian frequency of the applied field. For electrons whose energy exceeds the field photon energy, these calculations give algebraically the same rate of energy gain by the electrons as the classical microwave theory, while for lower energy electrons the inverse bremsstrahlung rate is greater. For these calculations the atom in an electron-atom collision is characterized by an effective Z , approximately 0.3 for a typical atom. The inverse bremsstrahlung collision frequency is a function of the second power of the effective Z . As the ionizing cascade proceeds, the ions formed provide a higher Z ($Z = 1$), second body, for the inverse bremsstrahlung collisions. Calculations are being made to determine whether the electrons formed remain in the vicinity of the ions sufficiently long that they gain energy at the faster rate this larger effective Z provides and, in this way, lead to the lower breakdown threshold observed experimentally.

A phenomenological application of the inverse bremsstrahlung mechanism has been made by Wright¹⁰ in a paper in which he points out a significant difference between the microwave and optical frequency cases. At optical frequencies, an atom upon being raised to the first excited state by a collision with an electron can be rapidly excited to higher states and ionized by direct photoexcitation and photoionization processes. Microwave photons with energies of 10^{-5} to 10^{-6} eV are too small to cause such direct excitation, and the ionization must all result from collisions with electrons heated by the incident radiation. Calculations have been carried out viewing the stimulated bremsstrahlung and inverse bremsstrahlung processes as elements of a one-dimensional random walk of an electron along an energy axis. With the large energy increments involved in the inverse bremsstrahlung process, for typical atoms studied only ~ 10 absorption steps are necessary for an electron to excite or ionize an atom in a collision compared with the thousands of steps required in the microwave picture. With so few steps, fortuitous collisions could result in a more rapid ionization in the inverse bremsstrahlung picture even though the average rates as described above are the same. The calculations, however, show no enhancement of the net ionization rate for the large photon inverse bremsstrahlung model over the rate predicted by microwave theory unless the number of photons involved is of order 1, 2, or 3.

An experimental test of the effects of this photon increment energy transfer associated with optical frequency breakdown is being undertaken in which measurements will be made of the development of breakdown in cesium vapor under ruby and neodymium laser irradiation. An electron upon interacting with a ruby laser photon has sufficient energy to directly excite the first excited state of cesium, and subsequent photoexcitation and photoionization can lead to rapid ionization of the cesium. Under neodymium irradiation, however, an electron must interact with more than one photon before it has sufficient energy to excite the cesium. Measurements of the breakdown threshold of cesium vapor with ruby and neodymium laser irradiation are being undertaken to determine whether the photon unit energy transfer associated with inverse bremsstrahlung energy absorption leads to a markedly different breakdown threshold in the two cases. With argon or the other gases studied to date, the ratio of the number of photons absorbed for excitation is small. With either ruby or neodymium irradiation many collisions are involved, and the calculations indicate an energy gain rate and ionization in these gases similar to that calculated by microwave breakdown theory. For the cesium, however, the ratio of the number of photon absorptions required is two or more, and the inverse bremsstrahlung picture predicts a much greater difference between the two thresholds than the microwave breakdown model.

TWELVE-MONTH STATUS EVALUATION AND FUTURE PROGRAM

During the first twelve months of this contract, the following specific objectives have been accomplished:

- a. Studies of gas breakdown by optical frequency radiation have been carried out in argon, helium, neon, and air over the pressure range from atmospheric pressure to 2000 psi using the $.69\mu$ and 1.06μ radiation from high-intensity ruby and neodymium lasers, respectively. With both ruby and neodymium radiation, breakdown in air was observed to require the highest field strength with successively lower field strengths required for the breakdown in neon, helium, and argon. At low pressures with either ruby or neodymium laser irradiation, the breakdown threshold was observed to decrease with pressure varying approximately as $\frac{1}{\sqrt{P}}$.
- b. Measurements have been made of the attenuation of the incident giant laser pulse by the breakdown plasma. For beam intensities slightly above the breakdown threshold, it was observed with both ruby and neodymium radiation that more than half of the laser beam energy can be absorbed in the plasma produced by the breakdown and that over 90% attenuation of the laser beam can occur during the later portions of the giant pulse. Measurements of the attenuation of an optical beam by the breakdown plasma at times following the incident giant pulse have been carried out using the cw beam from a helium-neon laser and show that the same 90% absorption is present for times of the order of

milliseconds after the formation of the plasma. These measurements demonstrate an extremely long lifetime for the breakdown plasma and indicate that plasma losses in the afterglow are extremely small.

- c. Measurements have been made to examine the effects of diffusion-like losses on the breakdown threshold by varying the focal volume within which the breakdown is formed. With both ruby and neodymium radiation, the breakdown threshold for all of the gases studied is inversely related to the dimensions of the breakdown region; i.e., breakdown within small focal volumes requires a larger optical frequency electric field than is necessary for larger volumes. These measurements have been carried out from atmospheric pressure to 2000 psi, and at all pressures the same effect is noted. The volume dependent breakdown threshold implies that even at pressures as high as 2000 psi, diffusion-like losses play a significant role in the development of optical frequency breakdown and that the loss-free breakdown threshold lies at still lower electric field strengths.
- d. In the experiments with neodymium irradiation, for the larger focal volumes with both helium and argon a pronounced minimum has been observed in the breakdown electric field vs. pressure curves. With ruby irradiation no minimum is observed for helium and that for argon is less distinct and shifted to higher pressures.
- e. Using the breakdown data obtained with ruby and neodymium laser irradiation, the frequency dependence of the breakdown threshold has been evaluated. For either argon, helium, or air the lower frequency neodymium radiation gives a lower breakdown threshold than for ruby at low pressures. At high pressures the neodymium data approaches (helium) or crosses (argon) that for ruby as a result of the minimum observed with neodymium; for air the ratio of the neodymium to ruby breakdown threshold remains approximately constant over the pressure range studied.
- f. Theoretical studies have been carried out which show that existing classical models of the breakdown process are not adequate to explain the phenomena observed at optical frequencies. Multiple photon theories recently proposed are unable to predict the magnitude of the E field required for breakdown or the pressure and volume dependence obtained experimentally. Calculations of the inverse bremsstrahlung cascade theory of the breakdown process have been carried out for optical frequencies where the photon energy is greater than the classically calculated electron oscillation energy and show that an electron exchanges energy with the applied electromagnetic field in increments of the photon energy. This result differs in kind from that obtained using the classical microwave theory and offers an experimental test to examine the validity of the inverse bremsstrahlung model.

On the basis of these results, the following areas are outlined for investigation during the next contract period:

- a. The presence of diffusion-like losses associated with the development of breakdown by optical frequency radiation has been demonstrated in the experiments, but the origin and nature of these losses is not yet adequately understood. Existing theories of the optical frequency breakdown process have made no attempt to include the effects of diffusion-like losses, and in order to evaluate these theories, studies are planned to extend the measurements to focal dimensions such that these losses are no longer significant in the development of breakdown. Other forms of losses are undoubtedly present, and studies will also be made of these as they effect a breakdown. In particular, measurements will be made of the rate of expansion of the breakdown plasma in an effort to determine the blast wave kinetic transport losses associated with the optical frequency breakdown.
- b. The theoretical studies have shown that the photon unit energy absorption of the inverse bremsstrahlung model will give results substantially different from those predicted by the formulas of classical microwave theory only in the case where excitation or ionization of the gas atoms occurs in 1, 2, or 3 energy absorption steps. An experimental test of this photon increment energy transfer associated with optical frequency breakdown will be made by measurements of breakdown in cesium vapor under ruby and neodymium irradiation.
- c. A cascade theory of the development of breakdown requires the presence of initial electrons to start the multiplication process. Experimental studies are planned to examine the effects of initial ionization within the focal volume on the breakdown threshold to determine what portion of the breakdown time is associated with the production of initial ionization and the subsequent cascade multiplication.

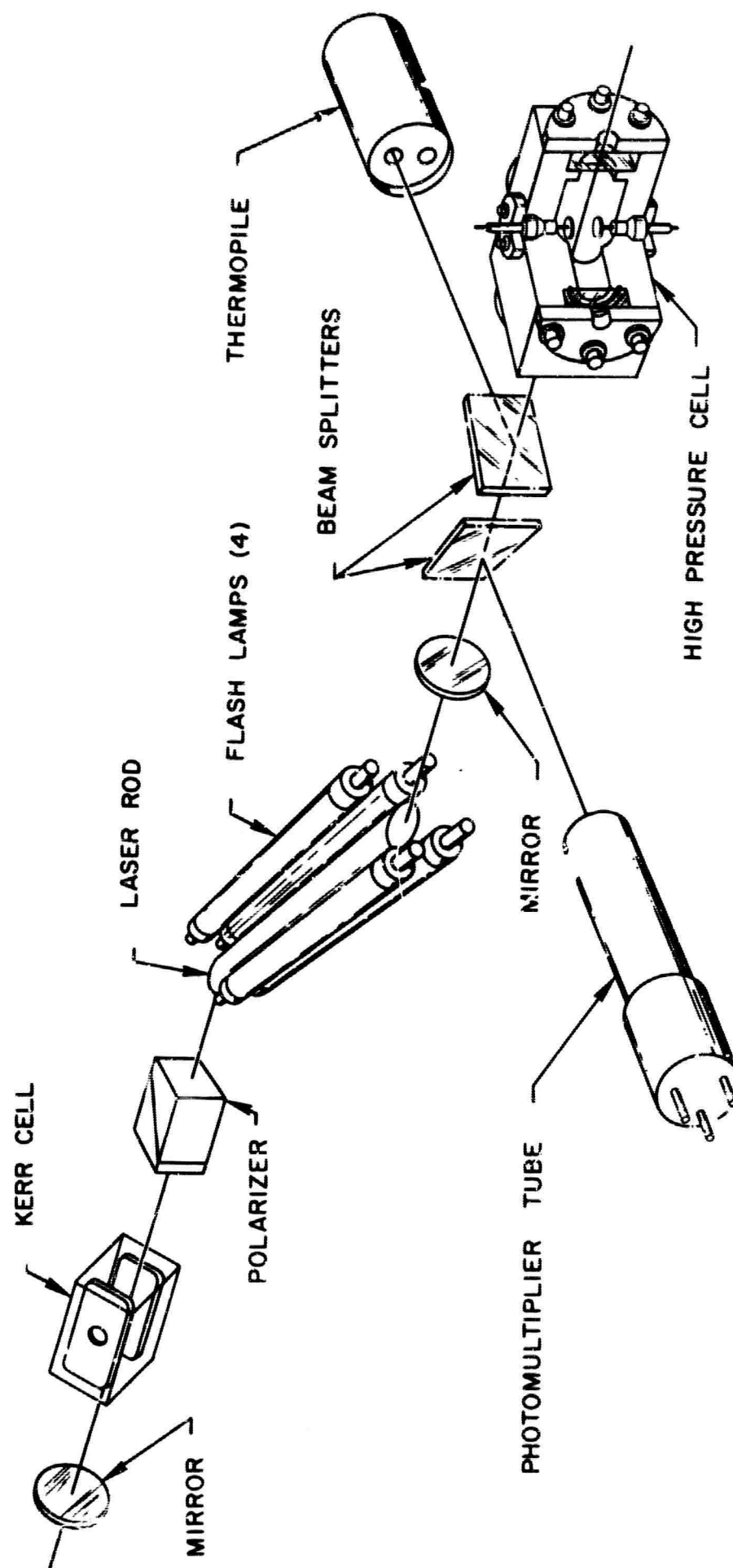
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LIST OF FIGURES

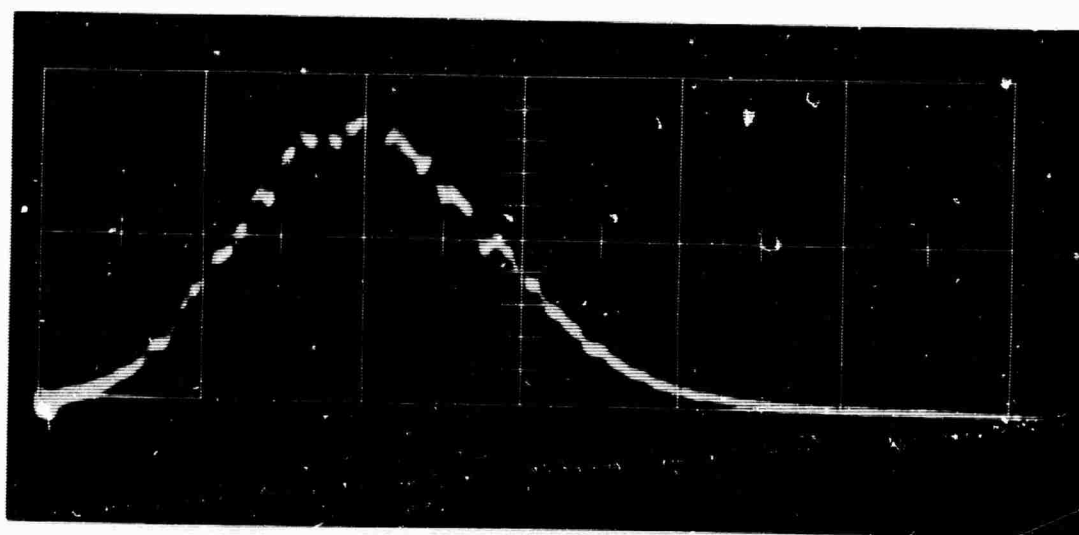
- Fig. 1 - Q-Spoiled Laser System
- Fig. 2 - Giant Pulse Wave Forms
- Fig. 3 - Breakdown Charge Collection
- Fig. 4 - Attenuation of Nd^{+3} Laser Beam by Breakdown Plasma
- Fig. 5 - He-Ne Beam Attenuation by Breakdown Plasma
- Fig. 6 - Breakdown vs. Pressure
- Fig. 7 - Breakdown vs. Diffusion Length
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- Fig. 10 - Breakdown vs. Frequency and Pressure

Q-SPOILED LASER SYSTEM



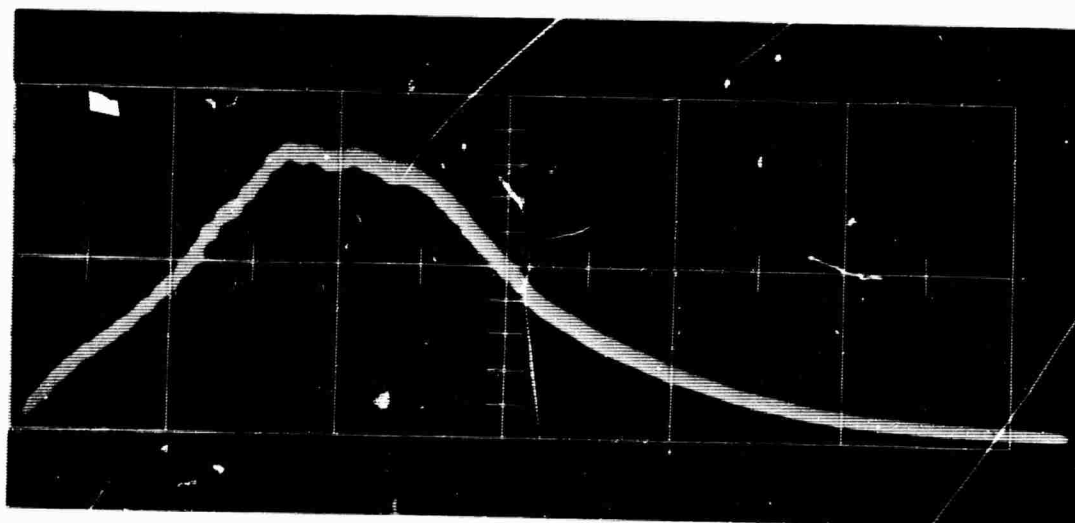
GIANT PULSE WAVE FORMS

RUBY
LASER
INTENSITY



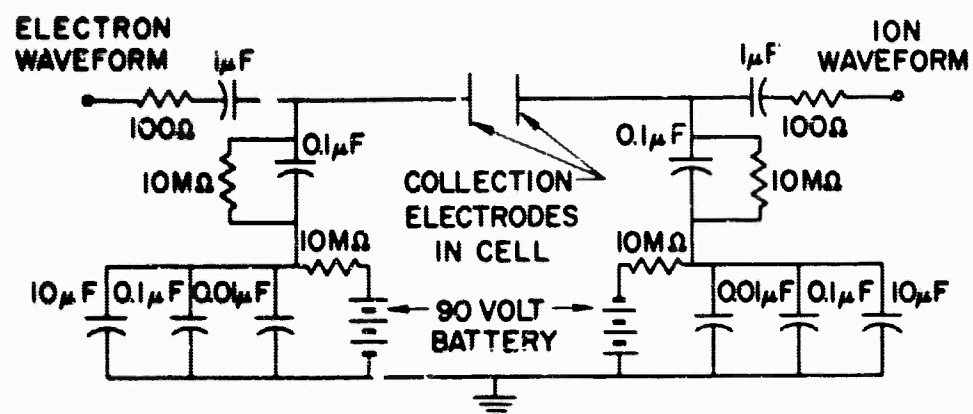
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NEODYMIUM
LASER
INTENSITY



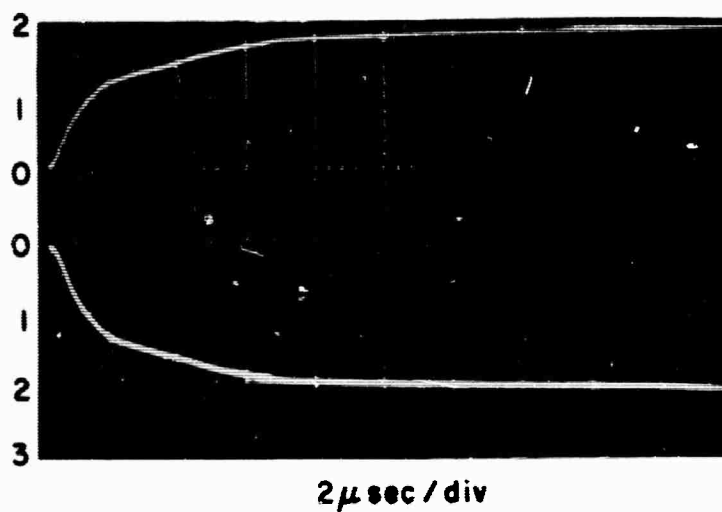
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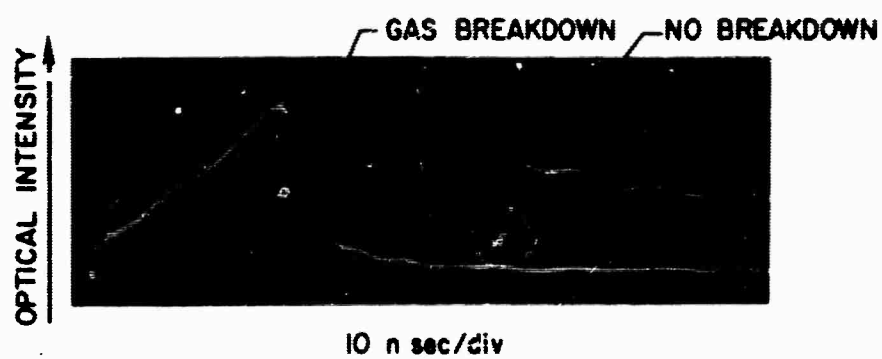
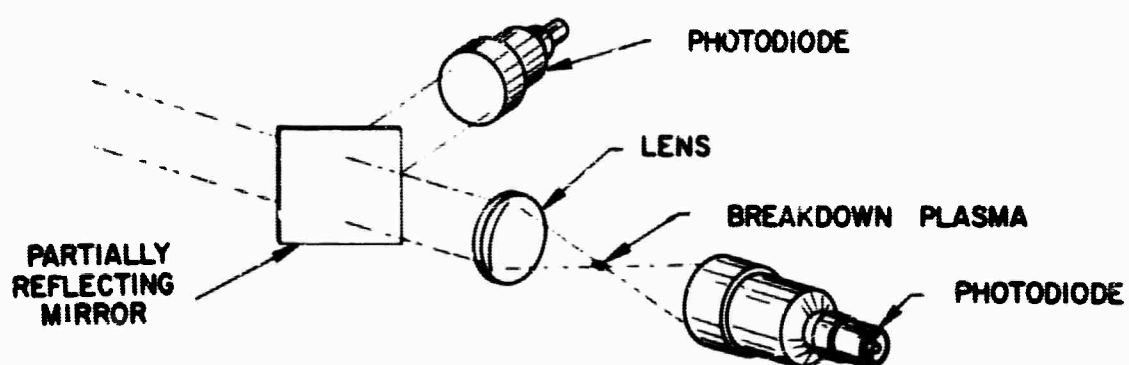
BREAKDOWN CHARGE COLLECTION



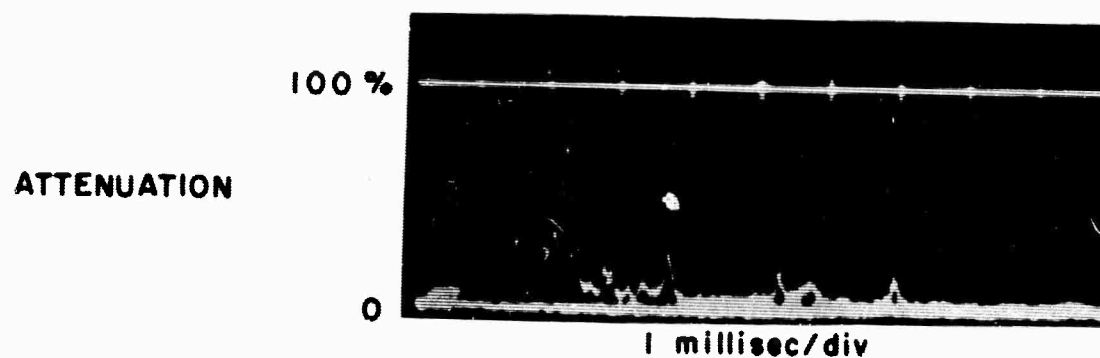
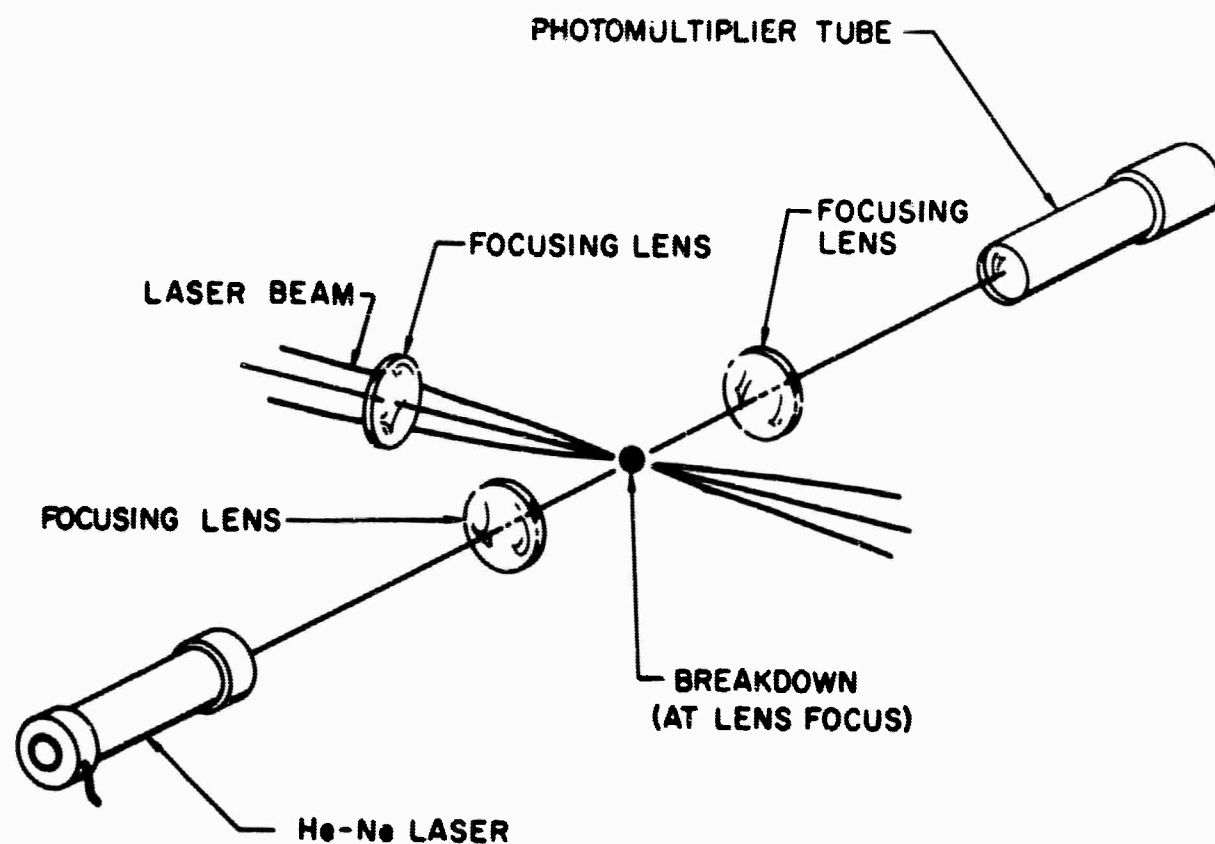
ION WAVE FORM
 0.6×10^{13} ions/div

ELECTRON WAVE FORM
 0.6×10^{13} electrons/div

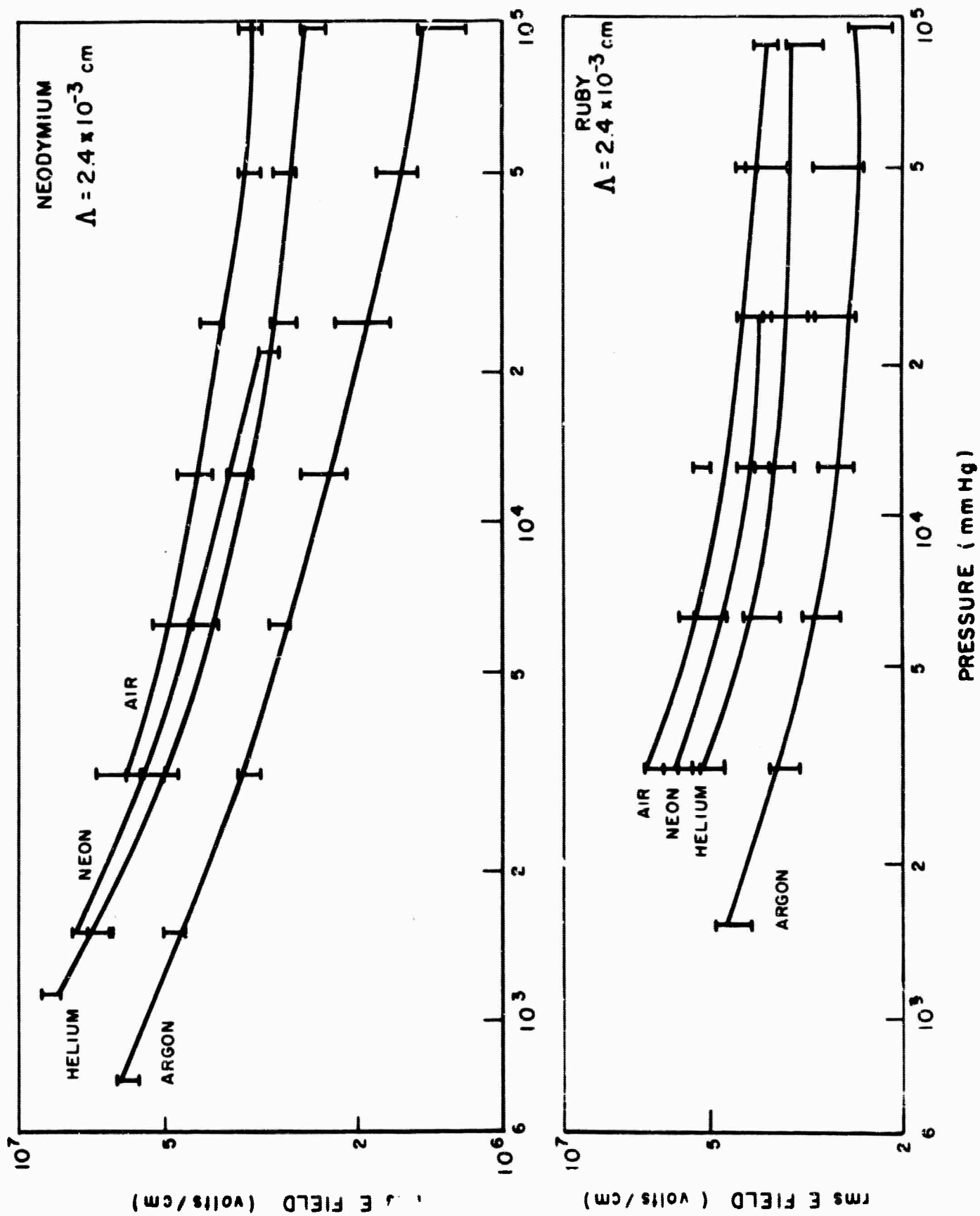


ATTENUATION OF Nd^{+3} LASER BEAM BY BREAKDOWN PLASMA

He-Ne BEAM ATTENUATION BY BREAKDOWN PLASMA



BREAKDOWN VS PRESSURE



BREAKDOWN VS DIFFUSION LENGTH

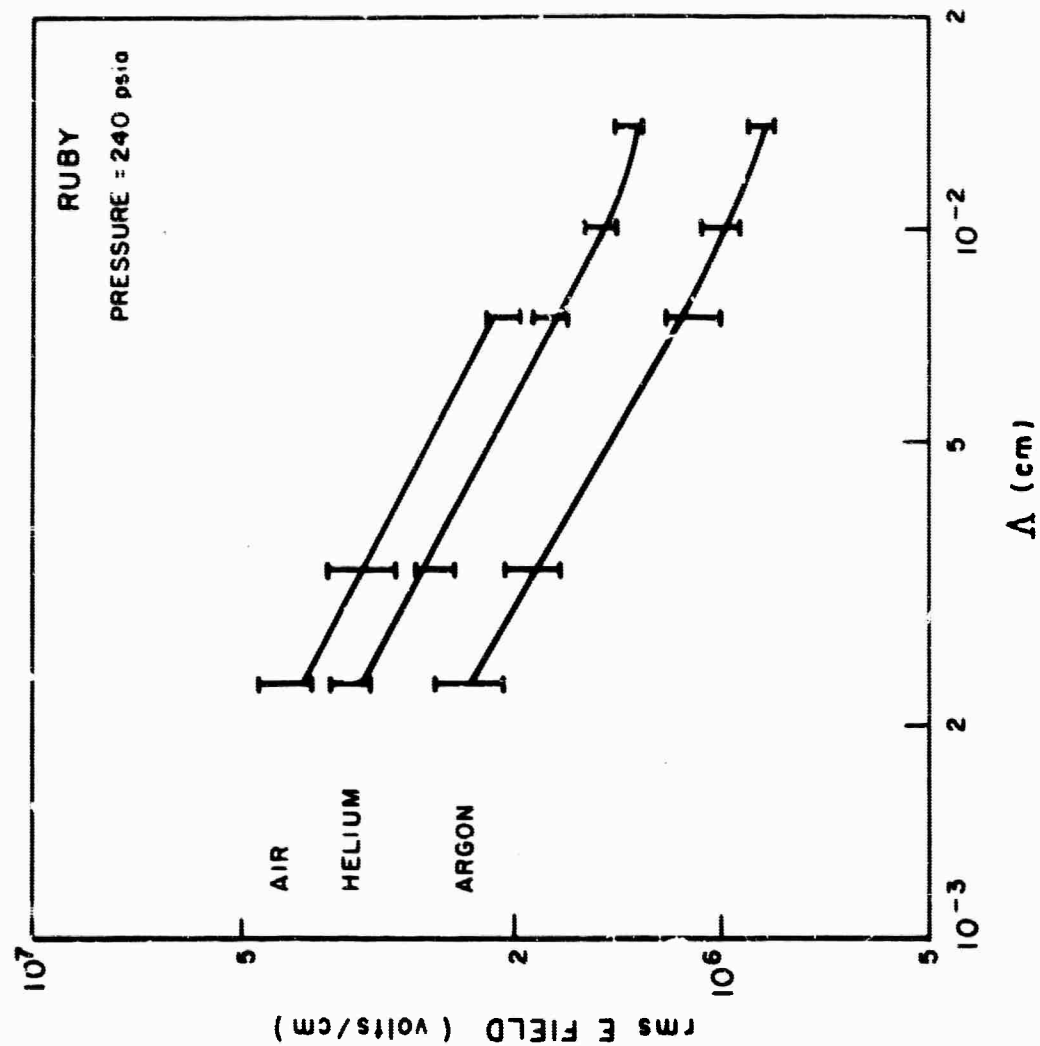
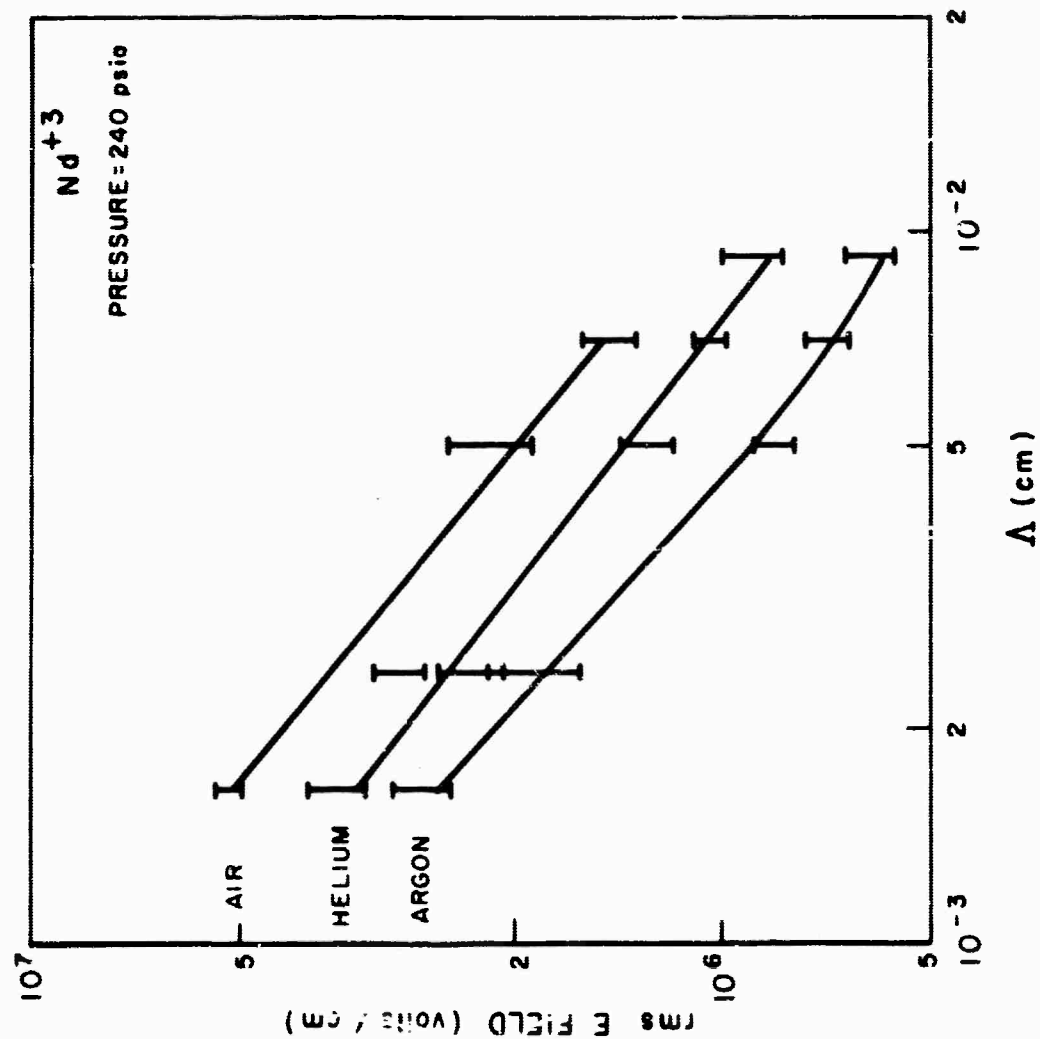
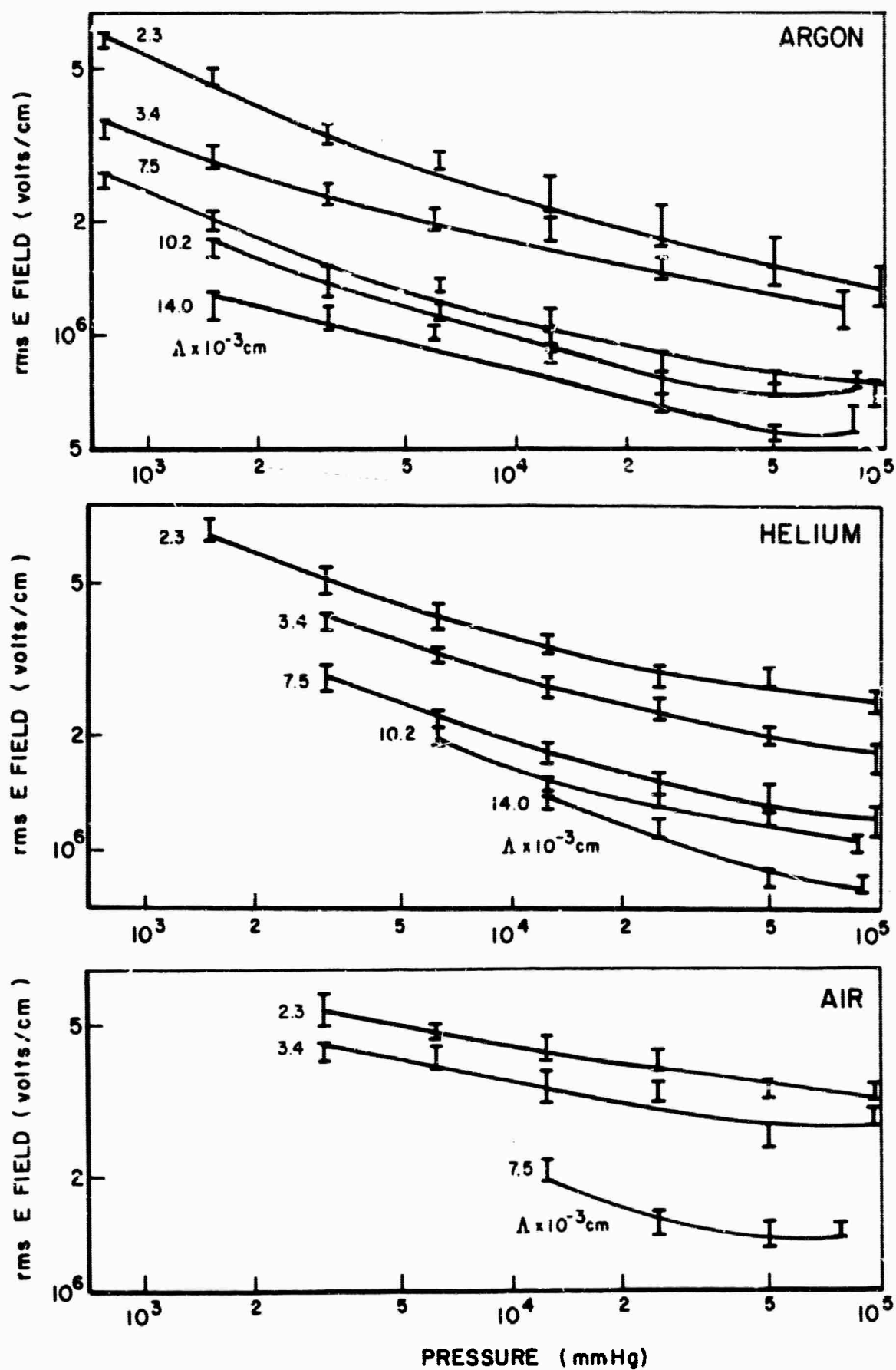
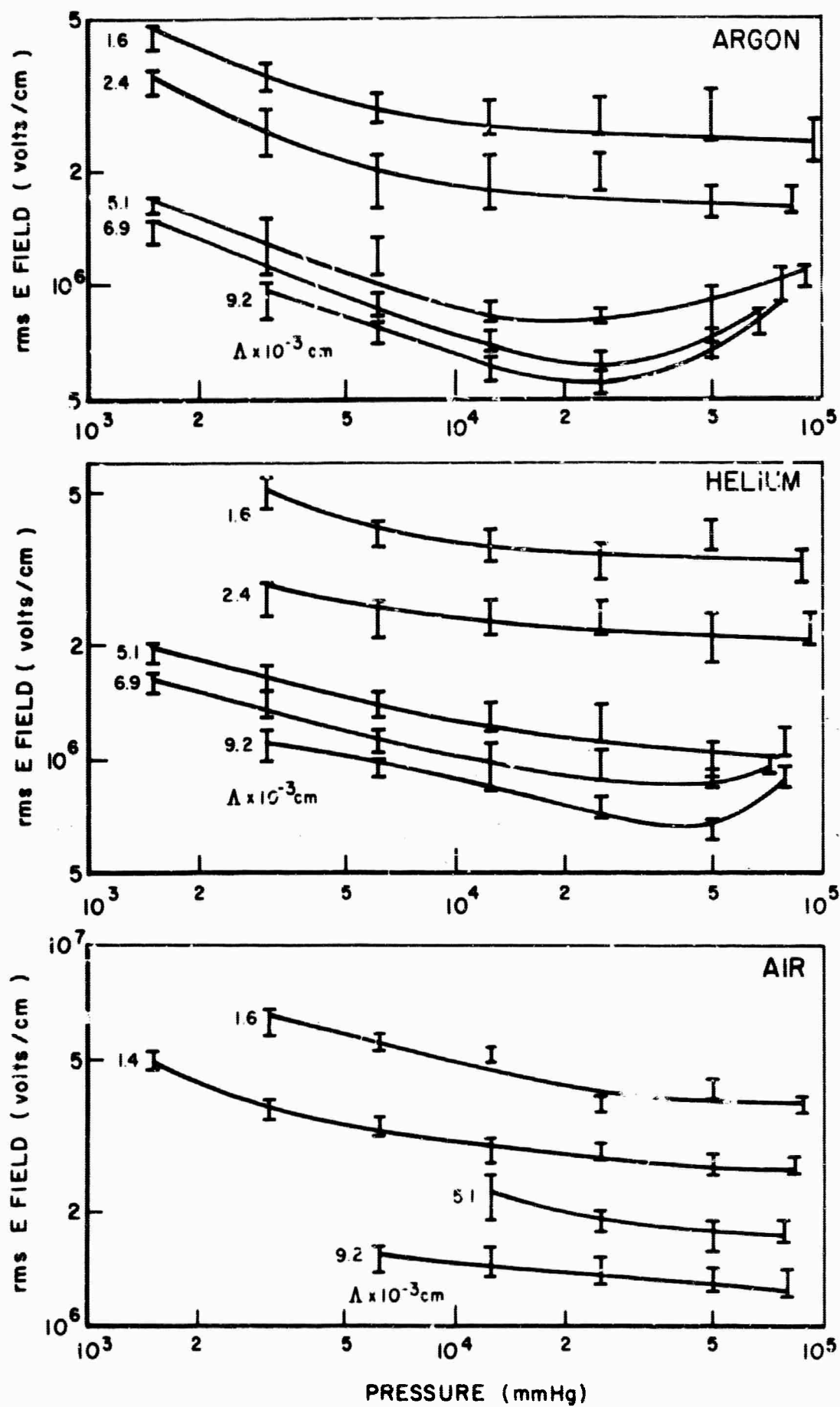


FIG. 7

BREAKDOWN VS PRESSURE AND Λ - RUBY

BREAKDOWN VS PRESSURE AND $\Lambda - \text{Nd}^{+3}$ 

BREAKDOWN VS FREQUENCY AND PRESSURE

